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Two-coefficient Cauchy model for low birefringence liquid crystals

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The three- and two-coefficient Cauchy equations are derived based on the three-band model for the wavelength- and temperature-dependent refractive indices of anisotropic liquid crystals. For high birefringence ($\Delta n \geqslant 0.2$) liquid crystals, the three-coefficient Cauchy model fits experimental results more accurately than the two-coefficient model. For low birefringence ($\Delta n \leqslant 0.12$) liquid crystal mixtures the two-coefficient Cauchy model works equally well as the three-coefficient model in the off-resonance spectral region. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1738526]

I. INTRODUCTION

Thin-film-transistor liquid crystal displays (TFT-LCDs) have been commonly used in notebook and desktop computers, cellular phones, and projection displays.[1,2] The fundamental light modulation mechanism of TFT-LCD is electric field induced liquid crystal (LC) refractive index change. The refractive indices of a LC are mainly determined by the molecular structure, wavelength, and temperature. To achieve a full-color display three primary colors (red, green, and blue) are needed. It is essential to know the wavelength- and temperature-dependent refractive indices of the LC mixture employed in order to optimize the cell design.

Several models have been developed to describe the wavelength and temperature dependencies of the LC refractive indices.[3–8] Each model has its own merits and incompleteness. For instances, the Vuks model[3] correlates the microscopic LC molecular polarizability to the macroscopic refractive indices. However, the wavelength and temperature effects are not described explicitly. The single band model[5,6] gives an explicit expression on the wavelength and temperature dependence for birefringence, but not for the individual refractive indices. The three-band model[7] describes the origins of the LC refractive indices for single LC compounds but requires three fitting parameters for each LC compound. If an LC mixture consists of a dozen different molecular structures, it would be too complicated for the three-band model to quantitatively describe the LC refractive indices of the mixture.

Although the original Cauchy equation[9] was intended for the isotropic gases and liquids, it has been attempted to fit the wavelength-dependent refractive indices of some anisotropic liquid crystals.[10,11] The fitting results are reasonably good except that the physical origins of the Cauchy coefficients are not clear. Recently, the extended Cauchy equations[12] were derived for anisotropic liquid crystals based on the Vuks model. The extended Cauchy equations are applicable not only to single compounds but also to LC mixtures. Good fittings are found in the off-resonance region. However, there are three Cauchy coefficients involved for each refractive index. It is highly desirable to reduce the numbers of the fitting parameters.

In this article, we derive a two-coefficient Cauchy model for the refractive indices of low birefringence LC mixtures. We find that if the LC birefringence is below 0.12 the three-coefficient Cauchy equations can be reduced to two coefficients. In Sec. II, we show the derivation processes for the two-coefficient Cauchy equations. In Sec. III, the experimental method for measuring the refractive indices is briefly described. In Sec. IV, we validate the two-coefficient Cauchy equations by fitting the experimental data of two low birefringence TFT liquid crystal mixtures: one with positive dielectric anisotropy ($\Delta\epsilon$) and the other with negative $\Delta\epsilon$. Excellent agreement between theory and experiment is obtained.

II. THEORY

In the three-band model, the refractive indices (n_e and n_o) are expressed as follows:[7]

$$n_e \cong 1 + g_{0e}\frac{\lambda^2\lambda_0^2}{\lambda^2-\lambda_0^2} + g_{1e}\frac{\lambda^2\lambda_1^2}{\lambda^2-\lambda_1^2} + g_{2e}\frac{\lambda^2\lambda_2^2}{\lambda^2-\lambda_2^2}, \tag{1a}$$

$$n_o \cong 1 + g_{0o}\frac{\lambda^2\lambda_0^2}{\lambda^2-\lambda_0^2} + g_{1o}\frac{\lambda^2\lambda_1^2}{\lambda^2-\lambda_1^2} + g_{2o}\frac{\lambda^2\lambda_2^2}{\lambda^2-\lambda_2^2}. \tag{1b}$$

Here, λ_0, λ_1, and λ_2 (with $\lambda_2 > \lambda_1$) denote the resonance wavelengths of the $\sigma \rightarrow \sigma^*$ and two $\pi \rightarrow \pi^*$ transitions, and g_0, g_1, and g_2 are the corresponding proportionality constants that depend on the oscillator strength and temperature. For a conjugated LC molecule, its λ_0 band is located in the vacuum ultraviolet region ($\lambda_0 \sim 120$ nm), λ_1 is around 190–210 nm; not too sensitive to the LC structure, and λ_2 increases substantially as the molecular conjugation increases. For example, for the 4-cyano-4-n-pentyle-cyclohexane-phenyl (5PCH) LC compound, its $\lambda_1 \sim 200$ nm and $\lambda_2 \sim 235$ nm while for the 4-cyano-4-n-pentylbiphenyl (5CB) its λ_1 shifts to 210 nm and λ_2 shifts to 282 nm.[7]

Equation (1) can be rewritten as follows:

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$$n_e \cong 1 + g_{0e}\frac{\lambda_0^2}{1-\left(\frac{\lambda_0}{\lambda}\right)^2} + g_{1e}\frac{\lambda_1^2}{1-\left(\frac{\lambda_1}{\lambda}\right)^2} + g_{2e}\frac{\lambda_2^2}{1-\left(\frac{\lambda_2}{\lambda}\right)^2}, \tag{2a}$$

$$n_o \cong 1 + g_{0o}\frac{\lambda_0^2}{1-\left(\frac{\lambda_0}{\lambda}\right)^2} + g_{1o}\frac{\lambda_1^2}{1-\left(\frac{\lambda_1}{\lambda}\right)^2} + g_{2o}\frac{\lambda_2^2}{1-\left(\frac{\lambda_2}{\lambda}\right)^2}. \tag{2b}$$

In the off-resonance region, $\lambda > \lambda_i$, $i=0$, 1, 2, the terms λ_i^2/λ^2 $(i=0,1,2)$ in the denominator are much smaller than 1 and Eq. (2) can be expanded into power series. Keeping the first three terms, we obtain

$$n_e \cong 1 + g_{0e}\lambda_0^2\left(1+\frac{\lambda_0^2}{\lambda^2}+\frac{\lambda_0^4}{\lambda^4}\right) + g_{1e}\lambda_1^2\left(1+\frac{\lambda_1^2}{\lambda^2}+\frac{\lambda_1^4}{\lambda^4}\right) + g_{2e}\lambda_2^2\left(1+\frac{\lambda_2^2}{\lambda^2}+\frac{\lambda_2^4}{\lambda^4}\right), \tag{3a}$$

$$n_o \cong 1 + g_{0o}\lambda_0^2\left(1+\frac{\lambda_0^2}{\lambda^2}+\frac{\lambda_0^4}{\lambda^4}\right) + g_{1o}\lambda_1^2\left(1+\frac{\lambda_1^2}{\lambda^2}+\frac{\lambda_1^4}{\lambda^4}\right) + g_{2o}\lambda_2^2\left(1+\frac{\lambda_2^2}{\lambda^2}+\frac{\lambda_2^4}{\lambda^4}\right). \tag{3b}$$

Grouping the similar terms together, we obtain the extended Cauchy equations

$$n_e \cong A_e + \frac{B_e}{\lambda^2} + \frac{C_e}{\lambda^4}, \tag{4a}$$

$$n_o \cong A_o + \frac{B_o}{\lambda^2} + \frac{C_o}{\lambda^4}, \tag{4b}$$

where

$$A_e = 1 + g_{0e}\lambda_0^2 + g_{1e}\lambda_1^2 + g_{2e}\lambda_2^2, \tag{5a}$$

$$B_e = g_{0e}\lambda_0^4 + g_{1e}\lambda_1^4 + g_{2e}\lambda_2^4, \tag{5b}$$

$$C_e = g_{0e}\lambda_0^6 + g_{1e}\lambda_1^6 + g_{2e}\lambda_2^6, \tag{5c}$$

and

$$A_o = 1 + g_{0o}\lambda_0^2 + g_{1o}\lambda_1^2 + g_{2o}\lambda_2^2, \tag{6a}$$

$$B_o = g_{0o}\lambda_0^4 + g_{1o}\lambda_1^4 + g_{2o}\lambda_2^4, \tag{6b}$$

$$C_e = g_{0o}\lambda_0^6 + g_{1o}\lambda_1^6 + g_{2o}\lambda_2^6. \tag{6c}$$

Therefore, the three-coefficient Cauchy equations are derived from the three-band model and are applicable to the anisotropic media. Each Cauchy coefficient is related to the resonance wavelengths (λ_i) and transition intensity (g_i) as shown in Eqs. (5) and (6). For instance, if a LC compound contains only σ electrons (e.g., cyclohexane rings), then λ_1 and λ_2 do not exist. The n_e and n_o are determined solely by the λ_o terms. As a result, the ABC coefficients would be small and the refractive indices in the visible region would be relatively small and insensitive to the wavelength. On the other hand, a linearly conjugated LC would exhibit a longer λ_2 and larger g_2 so that its refractive indices, especially birefringence $(\Delta n = n_e - n_o)$, would be greatly enhanced. From Eq. (4), both n_e and n_o decrease as the wavelength increases. In the long wavelength regime where $\lambda \gg \lambda_2$, n_e and n_o are reduced to A_e and A_o, respectively, and are insensitive to the wavelength. Hence, birefringence $(\Delta n = A_e - A_o)$ is also insensitive to the wavelength. This result is consistent with the prediction of the single band model.[5]

A LC mixture usually consists of several single compounds in order to widen the nematic range. Let us assume there are m compounds in the LC mixture and each compound contributes a molar fraction X_i (i=1,2,...m) to the refractive indices of the mixture. The refractive indices (n_{ei} and n_{oi}) of the ith component are expressed by Eq. (4). The refractive index of the mixture is a superposition of the individual components:

$$n_e \cong \sum_{i=1}^{m} X_i\left(A_{ei} + \frac{B_{ei}}{\lambda^2} + \frac{C_{ei}}{\lambda^4}\right), \tag{7a}$$

$$n_o \cong \sum_{i=1}^{m} X_i\left(A_{oi} + \frac{B_{oi}}{\lambda^2} + \frac{C_{oi}}{\lambda^4}\right), \tag{7b}$$

$$X_1 + X_2 + \cdots + X_m = 1. \tag{7c}$$

Let $A'_{e,o} = \sum_{i=1}^{m} X_i (A_{e,o})_i$, $B'_{e,o} = \sum_{i=1}^{m} X_i (B_{e,o})_i$, and $C'_{e,o} = \sum_{i=1}^{m} X_i (C_{e,o})_i$, Eqs. (7a) and (7b) are reduced to

$$n_e \cong A'_e + \frac{B'_e}{\lambda^2} + \frac{C'_e}{\lambda^4}, \tag{8a}$$

$$n_o \cong A'_o + \frac{B'_o}{\lambda^2} + \frac{C'_o}{\lambda^4}. \tag{8b}$$

Equation (8) represents the refractive indices of a LC mixture and has the same form as Eq. (4) which is for LC compounds, except for different Cauchy coefficients.

The three-coefficient Cauchy equations have been used to fit experimental results of LC mixtures. Good agreement is found in the off-resonance region.[12] An undesirable feature is that it involves three fitting parameters. To reduce the fitting parameters to two, we need to prove that the third terms, i.e., the λ^{-4} terms, in Eq. (8) can be ignored under certain conditions.

The fluorinated liquid crystals[13,14] exhibit a high resistivity, low viscosity, and excellent material stability, and have become the mainstream for direct-view and projection displays. For a 90° twisted nematic cell, to satisfy the Gooch–Tarry first minimum condition[15] the required $d\Delta n/\lambda$ is equal to $\sqrt{3}/2$. A cell gap $d \sim 4-5$ μm is commonly chosen in order to achieve high manufacturing yield. For the green band centered at $\lambda = 550$ nm, the LC birefringence should be in the vicinity of 0.1. The fluorinated cyclohexane phenyl has Δn in this range. The λ_2 band of such a LC structure occurs at $\sim$210 nm which is far from the visible (i.e., $\lambda_2 \ll \lambda$) and, moreover, its transition oscillator strength is weak,[16] which means the $g_{2e,o}$ coefficients are small.

From Eqs. (4a) and (5c), the C_e/λ^4 term of the single LC compound can be rewritten as

$$\frac{C_e}{\lambda^4}=g_{0e}\lambda_0^2\left(\frac{\lambda_0}{\lambda}\right)^4+g_{1e}\lambda_1^2\left(\frac{\lambda_1}{\lambda}\right)^4+g_{2e}\lambda_2^2\left(\frac{\lambda_2}{\lambda}\right)^4. \tag{9}$$

For a low birefringence LC compound in the off-resonance region, all the $(\lambda_i/\lambda)^4$ terms (where $i=0$, 1, and 2) in Eq. (9) are relatively small so that the C_e/λ^4 term can be ignored. Similarly, the third term for n_o as shown in Eq. (4b) can be neglected, too. As a consequence, the refractive indices of a low birefringence LC compound are reduced to

$$n_e \cong A_e+\frac{B_e}{\lambda^2}, \tag{10a}$$

$$n_o \cong A_o+\frac{B_o}{\lambda^2}. \tag{10b}$$

Based on the same arguments shown in Eq. (7), Eq. (10) holds equally well for low birefringence LC mixtures. From Eq. (10), there are only two Cauchy coefficients for each refractive index. If we measure the refractive index at two wavelengths, then the two Cauchy coefficients (A and B) can be obtained and the refractive indices at any wavelength can then be calculated. The A and B coefficients are related to each band through Eqs. (5) and (6). Their physical meanings are clear.

III. EXPERIMENT

We measured the refractive indices of 5CB, 5PCH, a commercial low birefringence fluorinated TFT LC mixture (Merck MLC-6241-000; $\Delta\epsilon=5.4$), and a fluorinated low birefringence negative ($\Delta\epsilon=-3$) LC mixture (UCF-280) using a multiwavelength Abbe refractometer (Atago DR-M4) at $\lambda=450$, 486, 546, 589, 633, and 656 nm. For some LC materials studied, their n_e or n_o is outside the measurement range at $\lambda=450$ and 486 nm. Thus, we will have fewer experimental data. The accuracy of the Abbe refractometer is up to the fourth decimal. For a given wavelength, we measured the refractive indices of 5CB, 5PCH, MLC-6241-000, and UCF-280 from 10 to 55 °C. The temperature of the Abbe refractometer is controlled by a circulating constant temperature bath (Atago Model 60-C3).

To find the upper boundary of the two-coefficient Cauchy model [Eq. (10)], we compare its fitting results with the three-coefficient Cauchy model using the experimental data of 5CB, 5PCH, UCF-280, and MLC-6241-000. The data of 5CB and 5PCH are taken from Ref. 7 because these two compounds have the most complete experimental refractive index data. At a given temperature, there are more than 50 refractive index data measured spanning in the 400–800 nm spectral range.

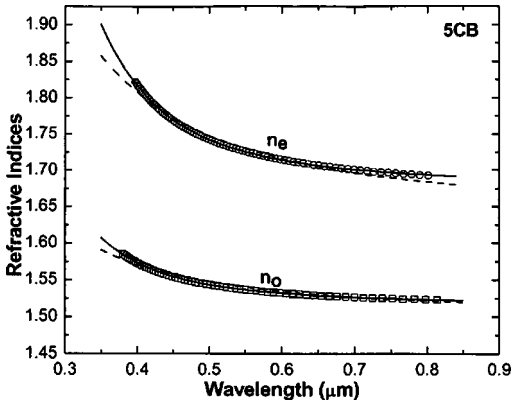

FIG. 1. Wavelength-dependent refractive index of 5CB at $T=25.1$ °C. Open circles and squares are experimental data for n_e and n_o, respectively. Solid lines are fittings using the three-coefficient Cauchy model [Eq. (4)] and dashed lines are fittings using the two-coefficient Cauchy model [Eq. (10)]. The fitting parameters are listed in Table I.

IV. RESULTS AND DISCUSSIONS

A. 5CB

Figure 1 depicts the wavelength-dependent refractive indices of 5CB at $T=25.1$ °C. Dots are experimental data and solid lines are fitting results using the three-coefficient Cauchy model and dashed lines are for the two-coefficient Cauchy model. The fitting parameters for both models are listed in Table I. In the visible and near-infrared regions, the three-coefficient Cauchy model fits the experimental data very well. In Table I, the χ^2 deviation is defined as the sum of the squares of observed values minus expected values and then divided by the expected values. For the three-coefficient Cauchy model, χ^2 is small ($\sim 10^{-7}$) indicating the fitting agrees with experimental results very well. On the other hand, the two-coefficient Cauchy model has a much larger χ^2 deviation ($\sim 10^{-5}$). This is because 5CB has a high birefringence ($\Delta n \sim 0.2$) so that the contribution of the λ^{-4} term to the refractive indices is still relatively large and cannot be neglected. From Table I, the C/B ratio of the Cauchy coefficients is around 1/2 for both n_e and n_o. This indicates that the magnitude of the λ^{-4} term is still comparable to the

TABLE I. Fitting parameters for the three- and two-coefficient Cauchy models: (LC) 5CB at $T=25.1$ °C. The units of Cauchy's B and C coefficients are μm^2 and μm^4, respectively.

	n_e				n_o			
Model	A_e	B_e	C_e	χ^2	A_o	B_o	C_o	χ^2
Three-coefficient Cauchy model	1.6795	0.0048	0.0027	3.54×10^{-7}	1.5174	0.0022	0.0011	9.99×10^{-8}
Two-coefficient Cauchy model	1.6427	0.0263		2.00×10^{-5}	1.5152	0.0105		7.47×10^{-6}

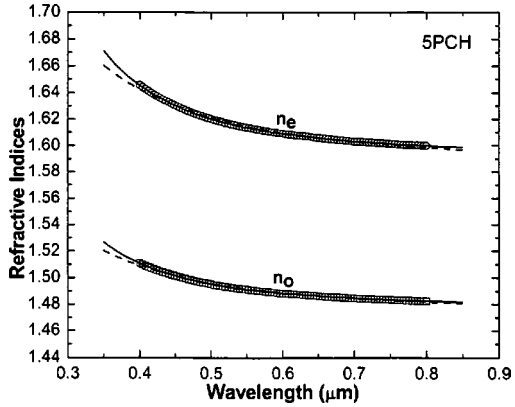

FIG. 2. Wavelength-dependent refractive indices of 5PCH at $T=25\,^{\circ}\mathrm{C}$. Open circles and squares are experimental data for n_e and n_o, respectively. Solid lines are fittings using the three-coefficient Cauchy model [Eq. (4)] and dashed lines are fittings using the two-coefficient Cauchy model [Eq. (10)]. The fitting parameters are listed in Table II.

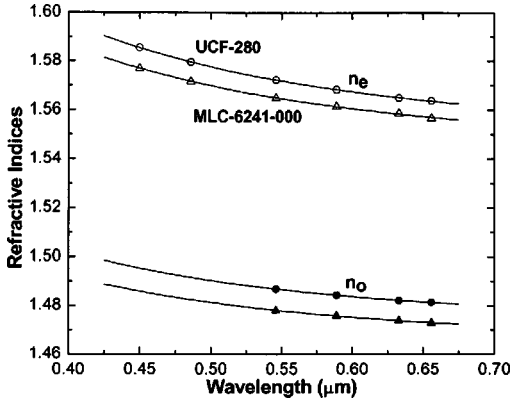

FIG. 3. Wavelength-dependent refractive indices of UCF-280 and MLC-6241-000 at $T=25\,^{\circ}\mathrm{C}$. Open and filled circles are experimental data for n_e and n_o of UCF-280, respectively. Open and filled triangles are experimental data for n_e and n_o of MLC-6241-000, respectively. Solid lines are fittings by using the two-coefficient Cauchy model [Eq. (10)]. The fitting parameters are listed in Table III.

λ^{-2} term and cannot be ignored. Therefore, for high birefringence ($\Delta n>0.2$) LC compounds and mixtures the three-coefficient Cauchy model should be used.

B. 5PCH

Figure 2 depicts the wavelength-dependent refractive indices of 5PCH at $T=25\,^{\circ}\mathrm{C}$. Dots are experimental data and solid lines are fitting results using the three-coefficient Cauchy model and dashed lines are for the two-coefficient Cauchy model. The fitting parameters for both models are listed in Table II. In the visible and near-infrared regions, the three-coefficient Cauchy model fits very well with the experimental data. The χ^2 deviation is as small as $\sim 10^{-9}$. On the other hand, the two-coefficient Cauchy model also fits the data well. Although its χ^2 deviation is still 2 orders of magnitude larger ($\sim 10^{-7}$), both deviations are indistinguishable. This is because 5PCH has a relatively small birefringence ($\Delta n\sim 0.12$) and the contribution of the λ^{-4} term to the refractive indices is negligible. From Table II, the C/B ratio of the Cauchy coefficients is $\sim 1/10$ for both n_e and n_o. This indicates that the λ^{-4} term is about one order of magnitude smaller than the λ^{-2} term. Therefore, $\Delta n\sim 0.12$ can be treated as the upper boundary that the two-coefficient Cauchy model begins to work as well as the three-coefficient Cauchy model.

C. Low birefringence LC mixtures

To validate that the two-coefficient Cauchy model is applicable to the refractive indices of the low birefringence LC materials, we chose two LC mixtures for this study: Merck MLC-6241-000 ($\Delta\epsilon>0$) and UCF-280 ($\Delta\epsilon<0$). The negative $\Delta\epsilon$ LCs are particularly useful for homeotropic alignment,[17] which exhibits an unprecedented contrast ratio. The refractive indices of these two mixtures were measured by our Abbe refractometer at $T=25\,^{\circ}\mathrm{C}$. Results are shown in Fig. 3.

In Fig. 3, circles and triangles represent the measured data for MLC-6241-000 and UCF-280, respectively, and solid lines are fitting results using the two-coefficient Cauchy model. The fitting parameters are listed in Table III. The χ^2 deviation is as small as $\sim 10^{-8}$. We also fit the experimental data by using the three-coefficient Cauchy model, the two curves overlap almost exactly. From Fig. 3, the birefringence of MLC-6241-000 and UCF-280 is found to be around 0.085 at $\lambda=550$ nm. Therefore, for low birefringence LC compounds and mixtures, the two-coefficient Cauchy model works quite well.

D. Temperature effect

Figure 4 depicts the temperature-dependent birefringence of 5CB, 5PCH, UCF-280, and MLC-6241-000 measured at $\lambda=589$ nm. The squares, circles, open triangles, and

TABLE II. Fitting parameters for the three- and two-coefficient Cauchy models: (LC) 5PCH at $T=25\,^{\circ}\mathrm{C}$. The units of Cauchy's B and C coefficients are $\mu\mathrm{m}^2$ and $\mu\mathrm{m}^4$, respectively.

	n_e				n_o			
Model	A_e	B_e	C_e	χ^2	A_o	B_o	C_o	χ^2
Three-coefficient Cauchy model	1.5903	0.0052	0.0006	5.13×10^{-9}	1.4763	0.0034	0.0003	1.59×10^{-9}
Two-coefficient Cauchy model	1.5838	0.0094		8.65×10^{-7}	1.4726	0.0058		2.98×10^{-7}

TABLE III. Fitting parameters for the two-coefficient Cauchy model: (LC) UCF-280 and MLC-6241-000 at $T=25\,^{\circ}\mathrm{C}$. The unit of Cauchy's B coefficients is $\mu\mathrm{m}^2$.

	n_e			n_o		
Materials	A_e	B_e	χ^2	A_o	B_o	χ^2
MLC-6241-000	1.5443	0.0083	1.51×10^{-8}	1.4689	0.0053	1.20×10^{-8}
	1.5395	0.0076	4.68×10^{-8}	1.4616	0.0049	9.69×10^{-9}

filled triangles are the measured birefringence of 5CB, 5PCH, UCF-280, and MLC-6241-000, respectively, at different temperatures. The temperature range is from 10 to 55 °C. At room temperature and $\lambda=589$ nm, the birefringence of 5CB, 5PCH, UCF-280, and MLC-6241-000 is 0.2, 0.12, 0.0839, and 0.0861, respectively. The solid lines represent fittings using the Haller equation[18]

$$\Delta n=(\Delta n)_o(1-T/T_c)^{\beta}, \tag{11}$$

where $(\Delta n)_o$ is the LC birefringence in the crystalline state, β is a material constant, and T_c is the clearing temperature of the LC material. The clearing point for 5CB, 5PCH, UCF-280, and MLC-6241-000 is 33.4, 52.9, 66.2, and 100 °C, respectively. From these fittings, we find $[(\Delta n)_o,\beta]=[0.3505,0.1889]$, $[0.1706,0.1512]$, $[0.1426,0.2513]$, and $[0.1221,0.2209]$ for 5CB, 5PCH, UCF-280, and MLC-6241-000, respectively. Although UCF-280 has a larger $(\Delta n)_o$ than MLC-6241-000, its clearing temperature is much lower. As a result, its birefringence at room temperature is lower than that of MLC-6241-000 due to the order parameter effect.

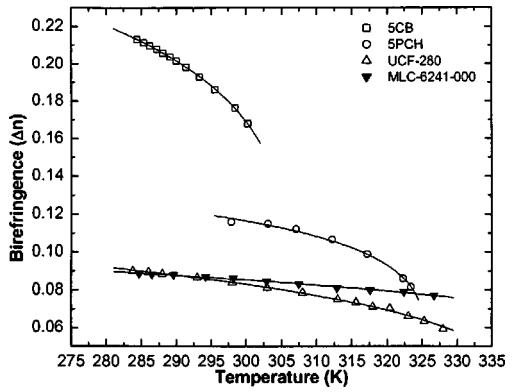

FIG. 4. Temperature-dependent birefringence (Δn) of 5CB (open squares), 5PCH (open circles), UCF-280 (open upward-triangles), and MLC-6241-000 (filled downward-triangles) at $\lambda=589$ nm. The four solid lines are fitting curves using $\Delta n=(\Delta n)_o(1-T/T_c)^{\beta}$, where T_c is clearing point. For 5CB: $(\Delta n)_o=0.3505$ and $\beta=0.1889$. For 5PCH: $(\Delta n)_o=0.1706$ and $\beta=0.1512$. For UCF-280: $(\Delta n)_o=0.1426$ and $\beta=0.2513$. For MLC-6241-000: $(\Delta n)_o=0.1221$ and $\beta=0.2209$.

V. CONCLUSIONS

We have derived and compared the three- and two-coefficient Cauchy models for describing the wavelength- and temperature-dependent refractive indices of LC compounds and mixtures based on the three-band model. If the LC birefringence is larger than 0.2, the three-coefficient Cauchy model has to be used. On the other hand, if the LC birefringence is smaller than ~0.12, the λ^{-4} term can be ignored and the two-coefficient Cauchy model works equally well as the three-coefficient Cauchy model. Most of TFT LC mixtures developed for direct-view and projection displays have a relatively low birefringence. Thus, the two-coefficient Cauchy model is adequate.

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